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SURVEY OF OZONE RESEARCH IN NORWAY

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1. OZONE RESEARCH BEFORE 1975

Ozone research has traditions in Norway. Observations of total ozone were started in 1939, and scientists interested in the climatology of atmospheric ozone are well acquainted with the long and nearly continuous record from Tromsö, situated at 70° North. Over a long time span this station provided important information about high latitude ozone. Shorter series of observations have been made at other stations: Dombås (62°N, 1940-1946) and Oslo (60°N, 1946-1949). Observations have also been made at Spitsbergen (79°N, 1950-1966). We have three Dobson photospectrometers at our disposal, but unfortunately none of these have been in operation since 1974. However, the instruments are now repaired and will be recalibrated. As soon as this process is finished, the instruments will be put in operation, one in Tromsö, one at Spitsbergen, and one in Oslo. (For details about ground based ozone measurements, see Appendix).

In addition to the ground based observations, the vertical distribution of ozone was measured in Oslo during the period 1968-1971 by means of ozone sondes. Altogether about 130 sondes were released. These observations constituted a part of the European ozone sonde network. However, the observations were abandoned

because of difficulties which arised from interference with local mobil-telephony. Further observations will depend on whether or not new radiocommunication equipment can be obtained. The future of ozone soundings from Oslo is therefore a question of economic resources and of training of personel.

While ground based ozone observations have long traditions ir Norway, theoretical work, in the sense we use this notation today, only covers a period of about ten years. Furthermore, only few people have been involved.

Photochemistry of the upper atmosphere was taken up as a research topic at the Institute of Geophysics, University of Oslo. The research has been directed by Professor Eigil Hesstvedt, and the size of the staff has varied from one to three. In addition, well qualified research work has been done by a number of students as a part of their Master's Degree.

In the beginning, the research was directed at chemical characteristics of the mesosphere and lower thermosphere. One of the more important results of this work was the introduction of vertical eddy transport in the photochemical models. This changed our picture of the vertical distribution of chemical components with long photochemical lifetimes. Today the importance of this effect is generally accepted, and all models of today include transport mechanisms in one, two or three dimensions. In the late 1960's, our research was centered around ozone in the lower stratosphere. After the importance of nitrogen oxides was demonstrated in 1971 independently by Johnston and Crutzen, our research followed a pattern similar to what we find in so many other institutes: a general study

of the chemistry of stratospheric ozone in order to evaluate the possible risk of a future fleet of high-flying aircraft. A series of papers were published about one- and two-dimentional modelling of the lower stratosphere. As an example, the first time-dependent photochemical model of the ozone layer was presented at the IAMAP General Assembly in Melbourne 1974. Much of this work fitted well into the goal of the Climatic Impact Assessment Program and was in part sponsored by the U.S. Department of Transportation. Our two-dimensional models were based on the best available knowledge of gas kinetics and stratospheric transport, and were used, among other things, to evaluate the effect of NO_x-injections from specified fleets and main routings of high flying aircraft (SST's). Figure 1 is taken from the Final Report (1974) of the CIAP and shows a comparison of various assessments of the SST effect.

2. OZONE RESEARCH AFTER 1975

Although we still shared an active interest in stratospheric problems, materialized in two institute reports, No.20 and 21 (see Appendix), our research has more and more become directed towards tropospheric ozone photochemistry. Our interest in tropospheric problems was initially raised a time after the successful oil drilling in the North Sea, when norwegian authorities were planning the development of petrochemical industry. In addition to inconveniences connected with industry in general, petrochemical industry involves emission of hydrocarbons and nitrogen oxides, which are the necessary ingrediences in photochemical smog formation. The analysis of the air pollution problems which could

arise in the surroundings of such industry was superficial, and it was felt that a better understanding of the mechanism of photochemical smog formation was needed. Against this background we decided to formulate a photochemical model which would cover the "clean" as well as the polluted atmosphere. A first version of the model was completed in the fall 1974. Later the model has been extended to include several different hydrocarbons which we observe in urban industrial areas.

Photochemical smog models is by no means a new invention; such models have been used in the analysis and forecasting of smog events. However, as a result of the complex dynamical and chemical character of the problem,, earlier models were based on very simplified chemical reaction scheme. In particular, chemistry of background air was simplified to a much larger extent than what is generally done in "clean" air modelling. The hydrocarbon chemistry often was eated by grouping reactions of the same type ("lumped" reactions). In contrast to this, our models make use of a "complete" background chemistry, i.e. we consider all known reactions which are likely to affect all the chemical components in the model. Furthermore, the hydrocarbon reaction scheme is complete in the sense that we consider explicitely the branching of the reaction scheme from the initial reactions of each hydrocarbon, through the intermediate steps to the end products.

It will appear from what we have said above that we may apply our model not only to simulate the chemistry of heavily polluted air, but also to describe the chemical composition of background ("clean") air and intermediate degrees of dilution which

occur when polluted air is transported away from its source and mixes with background air from the sides or from above.

I shall here describe briefly a model simulation of an atmospheric situation which serves as an example of present problem areas: photochemical oxidants, among them ozone, are known to occur in urban and industrialized regions almost all over the world. During the last years it has also become evident that these oxidants may in fact occur in high concentrations also when polluted air is transported into the environment and spred over large areas. The lifetime of ozone in such situations is long (of the order of days). To study such situations in more detail, we made the following model experiment:

We consider an air mass, with a prehistory of moderate emission rates, which is exposed to a somewhat stronger emissions of hydrocarbons and nitrogen oxides (HC/NO_x=1.5). After a period of six hours the emission is abolished, and the further chemical development in the air mass is determined by pure photochemistry in one case and combined with dilution through mixing with overlying "clean" air in another case. The dilution is quantified by an increase in the mixing height according to the equation $\Delta H=\sqrt{4Kt}$, corresponding to air transport over the sea. Our experiment shows tnat ozone continues to increase with a marked diurnal variation for about 24 hours after it left the source region. During this period NO_x is almost completely converted to HNO_3 while the hydrocarbons, with the exception of acetylene, are broken down. Later ozone increases slowly at a steady rate. The time constant for the ozone decrease will vary strongly from one situation to the next, depending on the emission of precur-

sors in the source region, vertical mixing and the rate of deposition on the underlying surface. However, 3 weeks may be regarded as indicative for the chemistry, while mixing with "clean" air will reduce this figure to 1 week or less. However, our experiment shows, and this is supported by measurements, that anthropogenic production of ozone in polluted areas will affect the ozone levels on a regional scale. This brings us to the point where our modelling becomes relevant for the problem we shall discuss during this conference: anthropogenic changes in ozone on a global scale. A rough estimate of the magnitude of the anthropogenic production of ozone on a regional scale can be made as follows: the averaged flux of NO_x over a large area (Central Europe) is taken to be 2.5×10^{11} molecules$\cdot$cm$^{-2}\cdot$s^{-1}. If we assume that each NO_x molecule on the average produces 2 ozone molecules (this is a low estimate) and a lifetime of one week, we arrive at an ozone increase of 3×10^{17} molecules$\cdot$cm^{-2} or 3% of the total ozone column density. This would apply to an area of the size of the industrialized part of a continent. A similar estimate of the increase in ozone averaged over the latitude belt 30-60°N results in 0.25%. In this case the anthropogenic production of ozone is comparable (~1/10) of the flux from the stratosphere.

Although these figures are relatively small, they are not insignificant. We feel that tropospheric ozone is not only an interesting topic in itself, but constitutes a part of the total ozone problem and as such deserves much attention in the future.

As was mentioned at the beginning of this survey, our research has also been directed towards stratospheric ozone.

Two reports were published last fall, one on a new method to compute the solar fluxes in the atmosphere and one, as a consequence of this, on the computation of more realistic values for photodissociation rate constants. The results of these two papers may be summarized as follows: Through the atmosphere, solar radiation is affected by a) absorption by atmospheric gases, b) molecular scattering, c) particle scattering, and d) reflection from clouds and from the ground. At the wavelengths of interest to stratospheric problems, O_2 and O_3 are the important absorbing gases. The reduction of solar fluxes due to absorption alone is easily taken care of when the total amount of O_2 and O_3 along the rays' path is known. The calculation is not very time consuming, and this effect is therefore included in the photochemical models where frequent recomputation of the fluxes due to a varying ozone distribution is necessary. On the other hand, the effect of multiple molecular (Rayleigh) scattering is more complex in its nature and therefore not so easy to quantify. Sundararaman et al. have solved the problem ir its complete form and found that the effect is considerable and cannot be ignored. However, the computation is too time consuming to be included in atmospheric models. In an institute report from 1974 (No.5) we proposed an approximate solution of the problem by simply assume that the solar beam, which is scattered in all directions, is affected in such a way that one half of the scattered light proceeds in the direction of the beam, the other half in the opposite direction. The method was worked out in more detail in a recent report (No.20, October 1976) and tested against the complete solution

by Sundararaman et al. (see Figure 2). In spite of the extreme simplification, our method appears to approximate the real effect of scattering in a most satisfactory way, and since it requires a reasonable computer time, it is now possible to include the effect of multiple molecular scattering in modelling work.

The solar fluxes establish the basis for computations of the photodissociation rate constants (J-values) of a number of gases we consider in our models. Hence, realistic J-values can only be obtained if our flux data are realistic. Since all computations of possible ozone reduction from high-flying aircraft or from increased release of fluorcarbons referred to in the report from the National Academy of Sciences (September 1976) have ignored the effect of multiple scattering, it is of interest to see how much the modified J-values affect the ozone depletion estimates. Two gases, NO_2 and $ClONO_2$, are of particular interest and the effect of including molecular scattering in the calculation of their photodissociation rate constants is demonstrated in Figures 3 and 4. We see that, in the stratosphere, the increase in solar fluxes due to multiple scattering results in a noticable rate increase. As an immediate result we derive that the stratospheric content of chlorine nitrate ($ClONO_2$) has been overestimated in previous model calculations. We have tested the magnitude of this error in a one-dimensional model and find (Sunde & Isaksen, Report No.21) that the results of previous calculations of the possible ozone reduction due to fluorcarbons are to be multiplied by a factor of about 1.4.

3. PLANS FOR FUTURE RESEARCH

As already mentioned ground based observations of ozon are planned to start again in a near future in Tromsö, Oslo, and Longyearbyen.

The theoretical modelling work at the Institute of Geophysics, University of Oslo, will continue.

The models we have developed are very general in their scope and may be used to study problems of different kinds. Reports of work on tropospheric ozone are in progress, and we have formulated further problems to be studied later. We are also preparing a two-dimensional model of the troposphere and stratosphere. Even this model may be adopted to various prob areas.

Our major concern is the financial situation. Funding for atmospheric ozone studies are small at present, and the terms for two of the three staff members expire in about a year's time. In view of the importance of this part of the global environmental problem I express the hope that groups working in this field in Norway will be given a fair chance t make use of its scientific research potential to contribute t the international co-operation on the ozone problem.

APPENDIX

OBSERVATIONS OF ATMOSPHERIC OZONE IN NORWAY

(Compiled by Dr. Søren H.H. Larsen)

TOTAL OZONE

Total ozone has been observed in Norway intermittently since 1939 at three stations: Tromsö (70°N), Dombås (62°N), and Oslo (60°N). Two Dobson spectrophotometers, "old type", No.8 and 14, were used. The earliest series of observations were published in Geophysica Norvegica: Tromsö (1939-49, Tönsberg), Dombås (1940-46, Langlo), and Oslo (1946-49). Both instruments were recalibrated and modernized in 1949/50. Instrument No.8 was brought to Longyearbyen, Spitsbergen (79°N), where observations started in 1950 (S. Larsen). Attempts were made to use the moon as light source for observations during the polar night. The results from 1950-51 are published in Geophysica Norvegica. Data for the period 1951-66 were sent to the ozone world data center. In 1967 the instrument was moved from Longyearbyen to Ny-Ålesund, Spitsbergen, where only sporadic series of observations were taken. The instrument collapsed in 1973. This instrument, No.8, belongs to the International Ozone Commission.

The observations in Tromsö (instrument No.14) were continued until 1974. Data for the period 1950-74 were sent to the ozone world data center. It was difficult to get competent operators to the instruments in Tromsö and at Spitsbergen, and since 1975 both instruments have been out of operation.

Norsk Polarinstitutt made a series of observations in Antartica (instrument No.56). The instrument was damaged and was left over to the Department of Physics, University of Oslo, where it has been repaired and calibrated against the standard instrument in Oxford.

The three instruments (No.8, 14, and 56) are now in Oslo. No.8 and No.14 are operational. No.14 will be sent back to

Tromsö where observations will be continued. Instrument No.8 will be sent to Longyearbyen where assistants at the meteorological office there will make the observations. No.56 will remain in Oslo, and new observations are planned. We also plan to make No.14 a standard instrument after intercomparison with another standard instrument.

VERTICAL DISTRIBUTION OF OZONE

Vertical distribution of ozone has been derived from optical ground-based observations in Tromsö (Tönsberg, Langlo) and at Spitsbergen (Larsen) (published in Geophysica Norvegica, 1945, and in Nature, 1954.

During the period 1968-71 a series of ozone sondes were flown from Oslo (Sören Larsen) under a project which aimed at the development of a combination of the MAST-sonde and the Väisäla meteorological sonde, which is used by the met. service in Norway. The results were satisfactory, but we had to make use of a Väisäla radioatively sonde to transmit the ozone signals. These sondes are not used by the Norwegian meteorological service. Observational data were exchanged with the European ozone sonde network and formed a part of the GARP experiment.

Difficulties with the telecommunications because of interference with mobil-telephoni has led to a temporary stop in the program. Our radiocommunication equipment has to be replaced in order to avoid interference. Ozone soundings require a well trained personal and considerable economic resources.

INSTITUTE REPORTS 1974-1977

INSTITUTE OF GEOPHYSICS (PHOTOCHEMISTRY GROUP)

UNIVERSITY OF OSLO

No.6 On NO_2-absorption of solar radiation and its role in the heat budget of the lower atmosphere (Hesstvedt, Isaksen,, August 1974)

No.8 Formation of oxidants and other secondary pollutants in air mixed with nitrogen oxides and ethylene (Hesstvedt, November 1974) (also in Geophysica Norvegica Vol.31, No.2)

No.9 Chemistry of a mixture of C_2H_4, NO_x, and air, studied in a simplified photochemical dispersion model (Hesstvedt, March 1975)

No.11 On the effect of chlorine on ozone formation in air polluted with C_2H_4 and NO_x (Hesstvedt, June 1975)

No.15 On the filtering effect of atmospheric ozone (Hesstvedt, February 1976)

No.16 On the chemistry of mixtures of hydrocarbons and nitrogen oxides in air (Hesstvedt, Hov, Isaksen, April 1976) (also in Geophysica Norvegica, Vol.31, No.6)

No.20 A simplified method to include molecular scattering and reflection in calculations of photon fluxes and photodissociation rates (Isaksen, Midtbö, Sunde, Crutzen, October 1976) (also in Geophysica Norvegica, Vol.31, No.5)

No.21 Stratospheric chlorine nitrate: reduced effect on the catalytical destruction of ozone due to molecular scattering and reflection of photon fluxes in the atmosphere (Sunde, Isaksen, November 1976)

No.24 Diurnal variations of ozone and other pollutants in an urban area (Hov, Isaksen, Hesstvedt, January 1977)

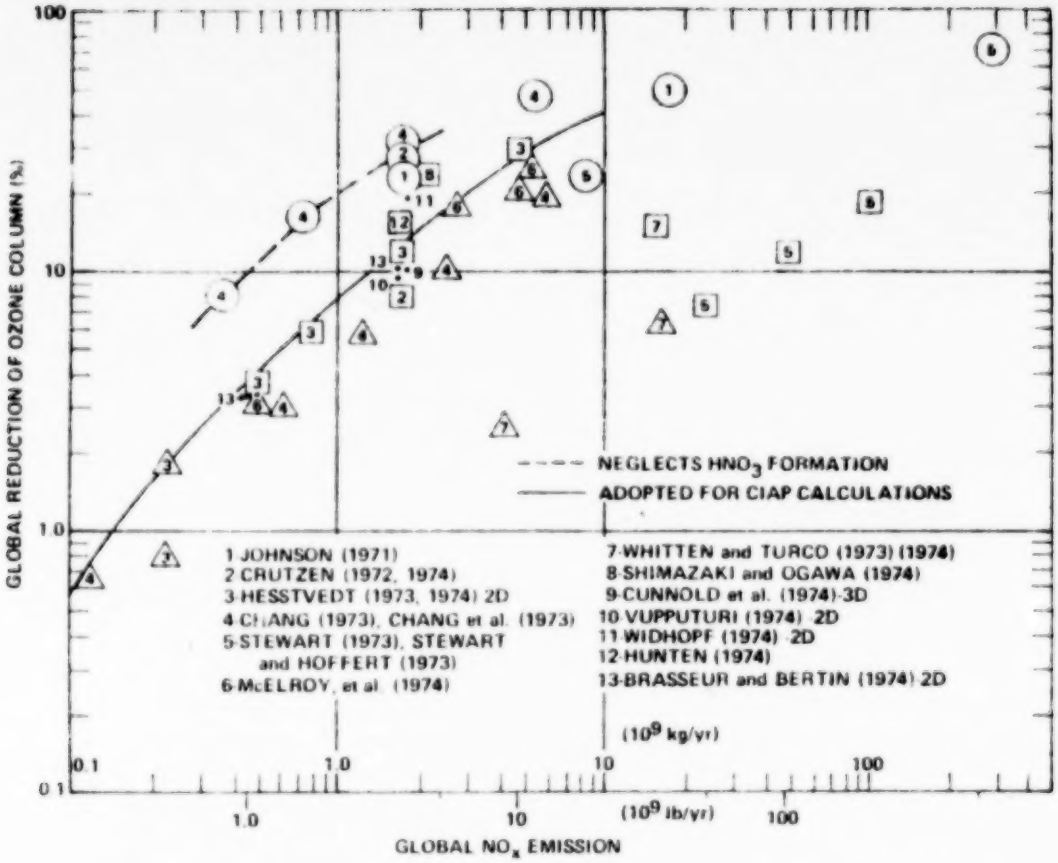

FIGURE 1. Calculated Reduction of Vertical Ozone in Terms of Uniform Global Rate of NO_x Addition at 20 km (66 kft)
Source: Chang and Johnston, 1974
(taken from CIAP, Final Report)

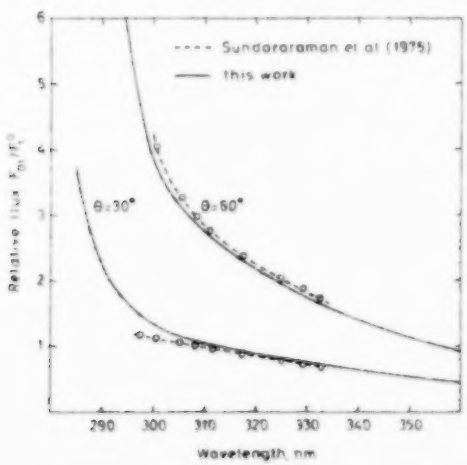

FIGURE 2. Comparison of complete and simplified computation of solar fluxes

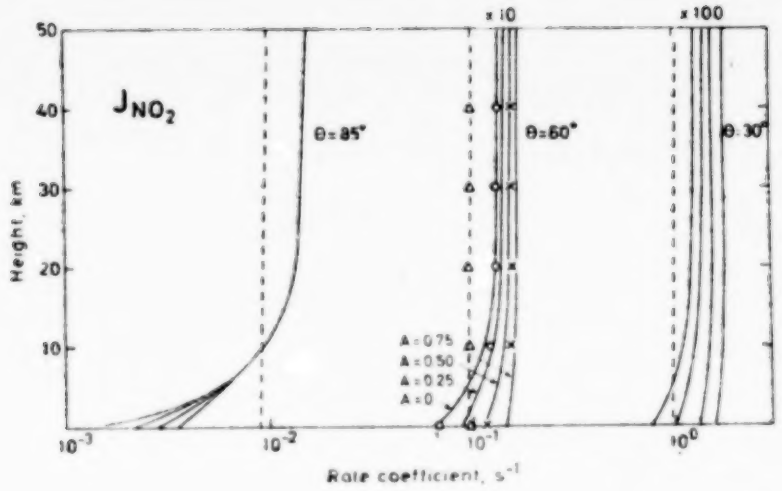

FIGURE 3. Rate constant for photodissociation of NO_2: broken line: pure absorption, solid line: absorption and multiple scattering

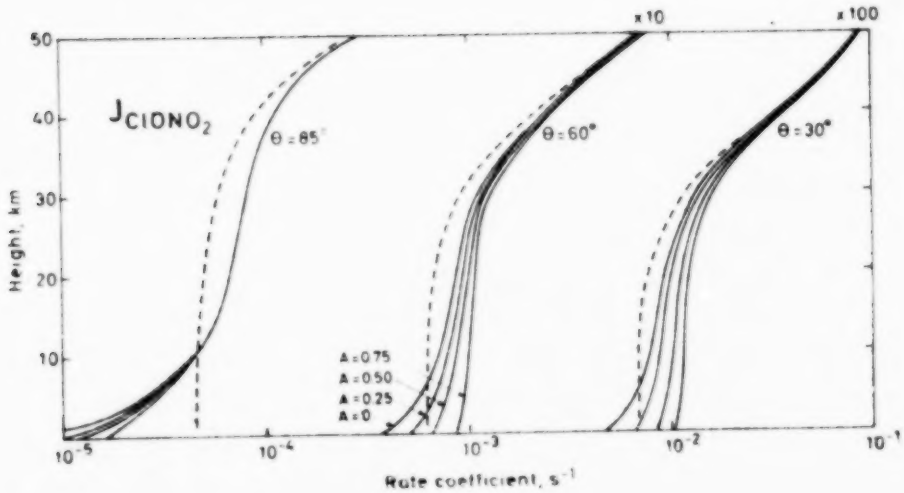

FIGURE 4. Rate constant for photodissociation of $ClONO_2$: broken line: pure absorption, solid line: absorption and multiple scattering